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Artificial Molecules: Antibonding Molecular Ground State for Holes Revealed

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Semiconductor quantum dots have long been described as artificial atoms because they have discrete energy states analogous to the energy levels of natural atoms. In recent years, it has become possible to create coupled pairs of quantum dots that are analogous to natural diatomic molecules. These artificial molecules have received a great deal of attention because of the potential applications in novel optoelectronic and spintronic devices, including the possibility of scalable implementations of quantum information processing. Just as in natural diatomic molecules, tunneling of electrons or holes between the two dots creates delocalized molecular orbitals. In natural diatomic molecules the molecular ground state has bonding orbital character and the first excited molecular state has antibonding character. Artificial quantum dot molecules were believed to behave in a similar way. However, recently Dr. Doty from the University of Delaware, Dr. Climente from Universitat Jaume I (Castellon, Spain), and their collaborators in Canada and the US, have experimentally verified and explained the existence of an antibonding molecular ground state for holes in artificial quantum dot molecules.

The coherent coupling of quantum dots leads to the formation of delocalized molecular orbitals that appear in photoluminescence spectra under electric fields as ant crossings. The orbital character of the molecular states cannot be measured at zero magnetic field. However, a recent discovery by Doty and coworkers at the Naval Research Lab revealed that, when a magnetic field was applied, the resonant changes in the Zeeman splitting that depended on the orbital character of the molecular states appeared. Using these changes to identify the molecular orbital character, Doty and coworkers demonstrated that the orbital character of the molecular ground state reverses as a function of the distance between the dots. Holes confined in the molecular states of dots separated by 2 nm have the expected bonding molecular ground state; when the dots are separated by three or more nanometers, the molecular ground state becomes antibonding. "This was a surprising discovery for us," said Doty. "Until we looked closely at the orbital character of the molecular states we had simply assumed that the molecular ground state was always a bonding orbital."

To explain this surprising result, Climente and coworkers utilized a four-band $k \cdot p$ approximation which shows that the parity along the molecular axis is broken by the spin-orbit interaction in the valence band. "This leads to the mixing of bonding and antibonding heavy- and light-hole components of the spinor which destabilizes (stabilizes) the otherwise pure bonding (antibonding) states, leading to the state reversal" Climente told Nanospotlight. He continued explaining that the molecular ground states are found to have up to $\sim 95\%$ antibonding character as a result of this mixing, "This is about 10 times higher than the largest value observed in natural molecules, so we can speak about a novel kind of molecular state whose properties are still to be explored".

A $sp^3d^5s^*$ tight binding multimillion atom calculation was applied to probe theoretically a real case. "We considered self-assembled InGaAs/GaAs double quantum dot structures and included strain, structural asymmetries as well as vertical electric fields. The results are in qualitative agreement with the $k \cdot p$ theory and predict a bonding-to-antibonding ground state reversal at interdot distances of $d \sim 2$ nm" says Climente.

This discovery drastically changed the previous conception of the single-particle ground state of holes in coupled quantum dots. "This paves the way for more accurate simulations of device performance for

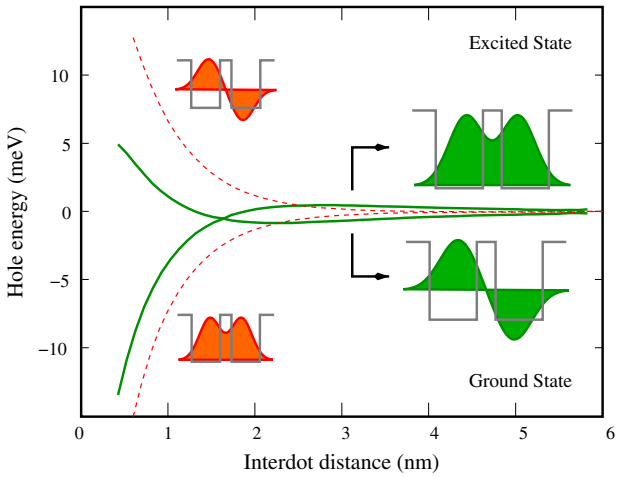

Fig. 1 Dissociation spectrum of a hole in an artificial molecule (solid line). Note the state reversal at $d \sim 1.5$ nm, implying a bonding-to-antibonding ground state transition. This never occurs for electrons in artificial or natural diatomic molecules, where the dissociation follows the pattern indicated by the dashed line. The insets are schematics of the molecular wavefunction orbital in the double quantum dot

applications in optics, transport, or quantum information" Climente mentioned. He continued saying that the tunneling rate, which is an important parameter for the quantum computation and transport can now be flexibly tuned from large values down to zero using a magnetic field. "This is a consequence of the spinor nature of holes" says Climente, "and it shows that we can greatly manipulate the properties of artificial molecules through the spin-orbit interaction". As a matter of fact, this discovery provides new tools for engineering the spatial distribution of molecular wavefunctions in regions with varying material parameters. "Wavefunction engineering can be used to control magnetic, spin, and optical properties, so this discovery will enable the design of wide variety of materials for novel optoelectronic applications," said Doty (Fig. 1).

This work is also featured in the OAtube nanotechnology journal, http://www.oatube.org/2008/09/jiclimente.html. This journal is a new kind of open access effort that offers video access to science.

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